High-resolution Larmor diffraction: theory and applications

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A.A. van Well, M.T. Rekveldt, J. Phys: Conf. Series **862** (2017) 012029

OUTLINE

- Introduction

  - why high-resolution diffraction
  - ‘conventional’ neutron diffractometers

- Larmor diffraction

  - double-arm geometry
  - examples

- Larmor diffraction

  - single-arm geometry
  - examples

- Discussion

HIGH-RESOLUTION NEUTRON DIFFRACTION

- Crystal-structure determination
  - powder diffraction with closely separated Bragg peaks
- Small lattice-parameter changes during (quantum) phase transitions [1]
- Weak magneto-elastic effect [2]
- Materials / engineering science [3,4]
  - precise value of lattice parameter
  - shape of Bragg peak

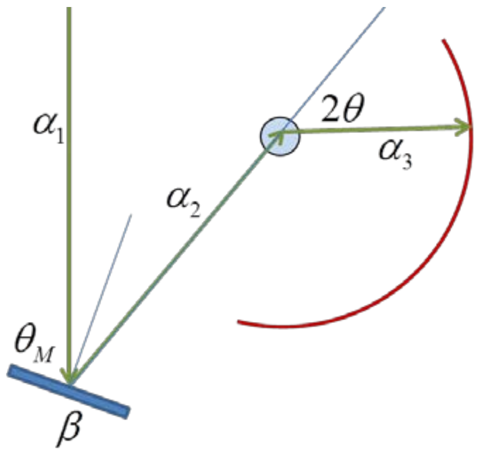
  → info about different types of microscopic stresses (defects / more phases)
  internal stress in large-Young's-modulus materials (ceramics)
  lattice misfits in multi-phase materials

[1] Pfleiderer et al., Science **316** (2007)1871
[2] Martin et al., J. Phys.: Conf. Series **340** (2012) 012012
[3] Lucks et al., Appl. Cryst. **37** (2004) 300
[4] Repper et al., Acta Materialia **58** (2010) 3459

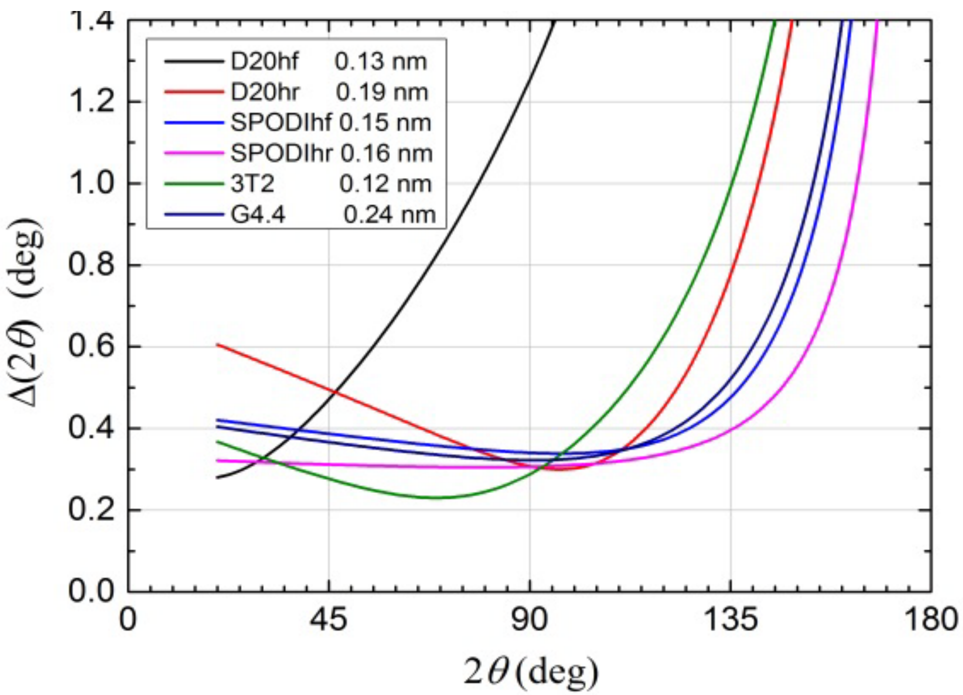
CONVENTIONAL DIFFRACTOMETERS

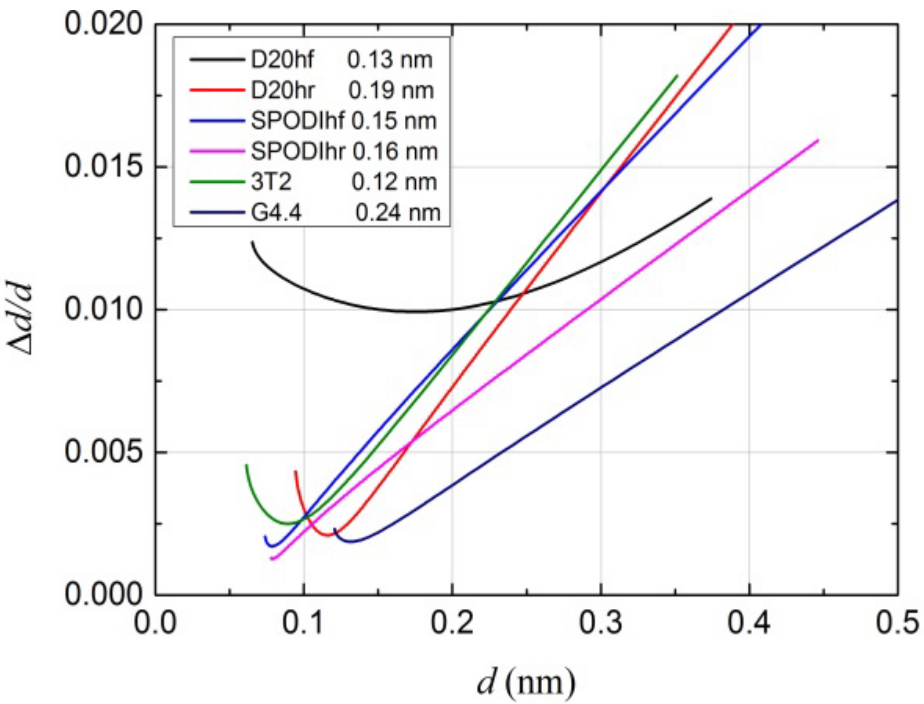
Constant-wavelength diffractometers (reactors)

- Resolution is highly scattering-angle dependent [1,2]
- High resolution $\Delta d/d \approx 1 \cdot 10^{-3}$ for limited d-spacing range



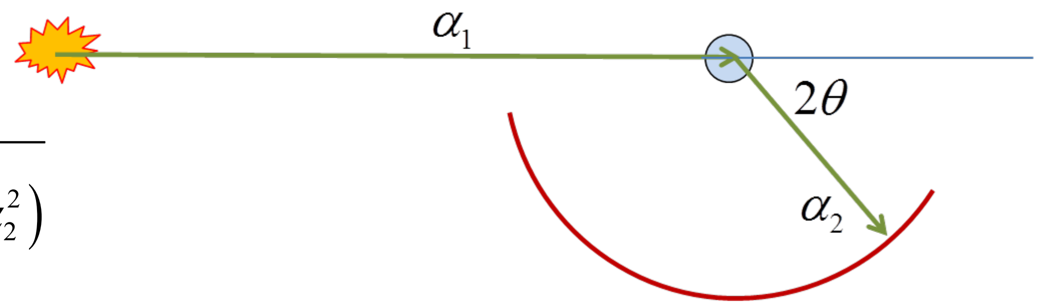
$$\Delta(2\theta) = \sqrt{W - \frac{V^2}{4U}} \qquad \frac{\Delta d}{d} = \cot\theta \; \Delta\theta$$



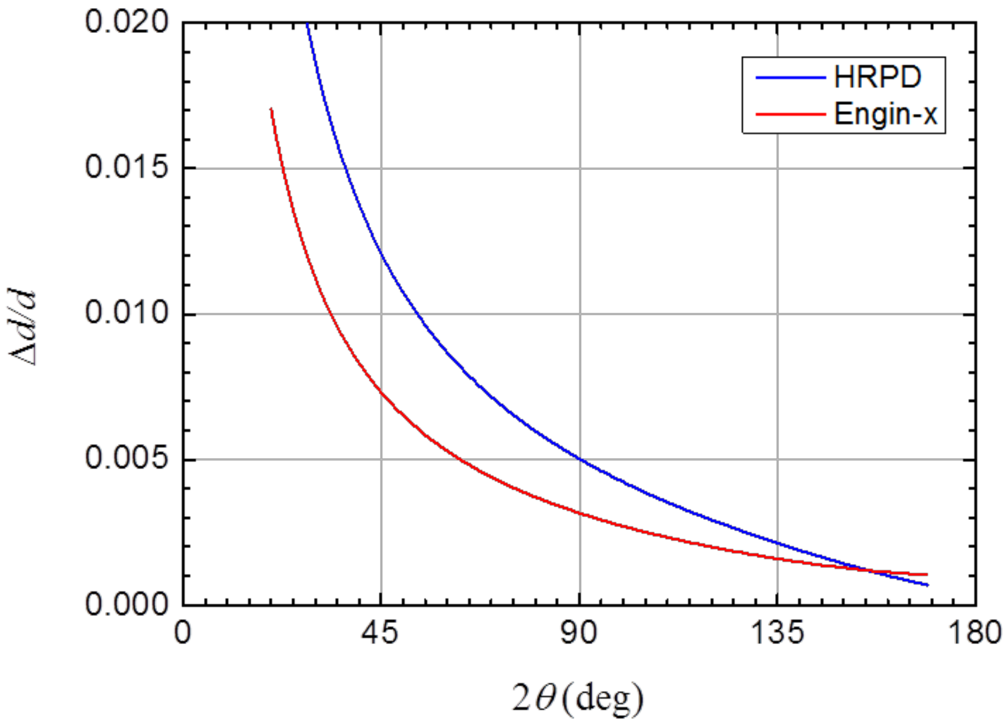


[1] Caglioti et al., NIM **3** (1958) 223
[2] Hewat et al., NIM **127** (1975) 361

CONVENTIONAL DIFFRACTOMETERS
Time-of-flight diffractometers (spallation sources)



$$\frac{\Delta d}{d} = \sqrt{\left(\frac{\Delta t}{t}\right)^2_{\mathrm{mod}} + \left(\frac{\Delta L}{L}\right)^2 + \frac{\cot^2\theta}{4}\left(\alpha_1^2 + \alpha_2^2\right)}$$

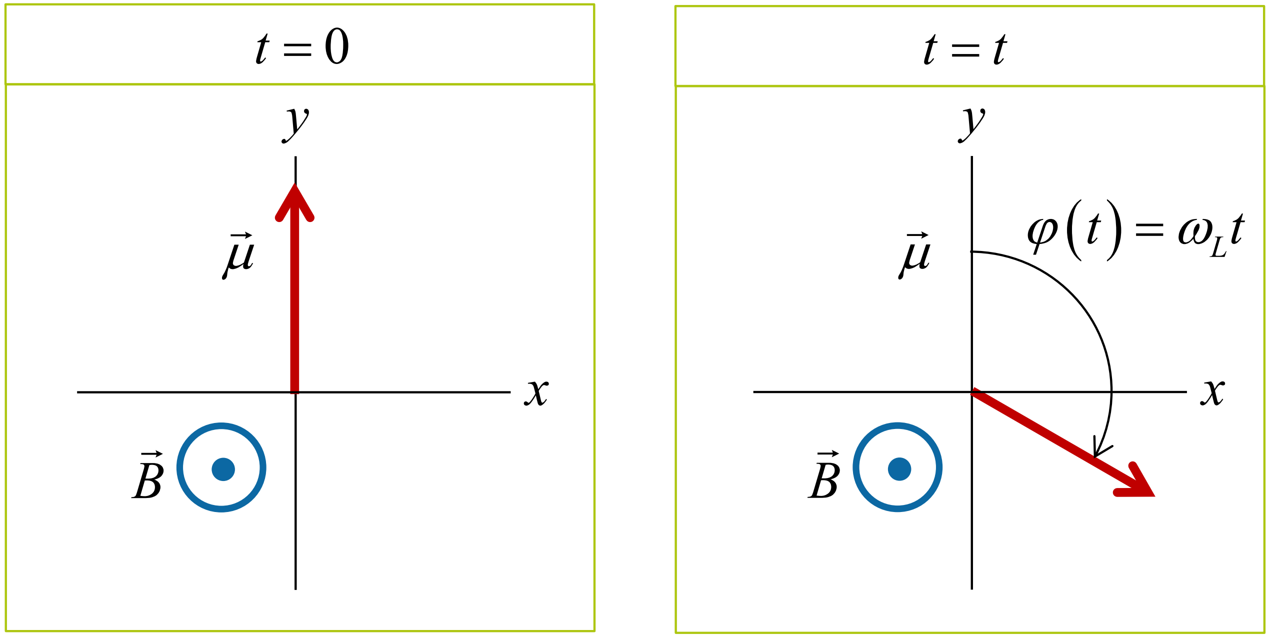


Q resp. d range depends on
- wavelength range
- and scattering angle

LARMOR DIFFRACTION

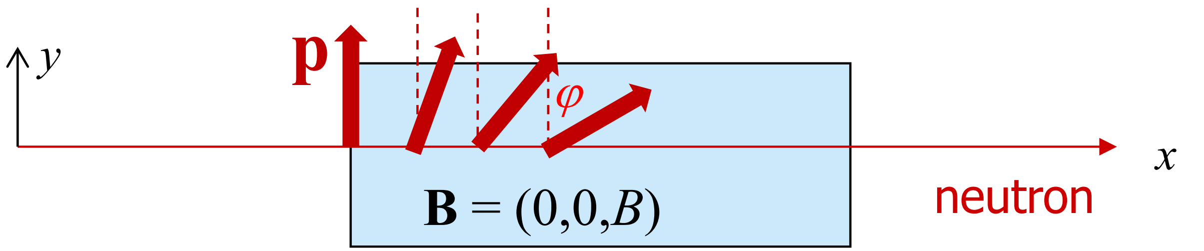
High resolution, decoupled from divergence and monochromaticity

How does it work? Using Larmor precession of polarised neutrons



polariser aligns neutrons in y-direction

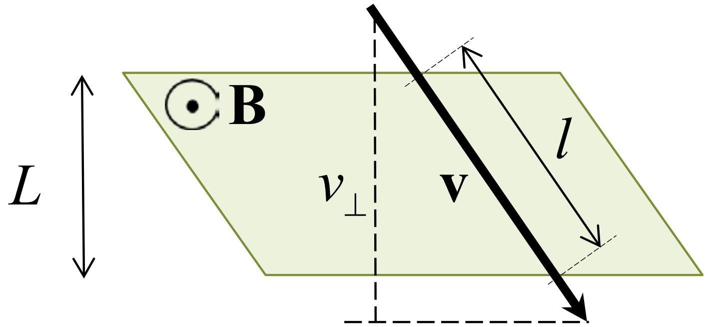
analyser measures <cos φ>



LARMOR DIFFRACTION

High resolution, decoupled from divergence and monochromaticity

Neutron with in-plane spin and velocity $\mathbf{v}$ transverse parallelogram-shaped perpendicular magnetic field $\mathbf{B}$

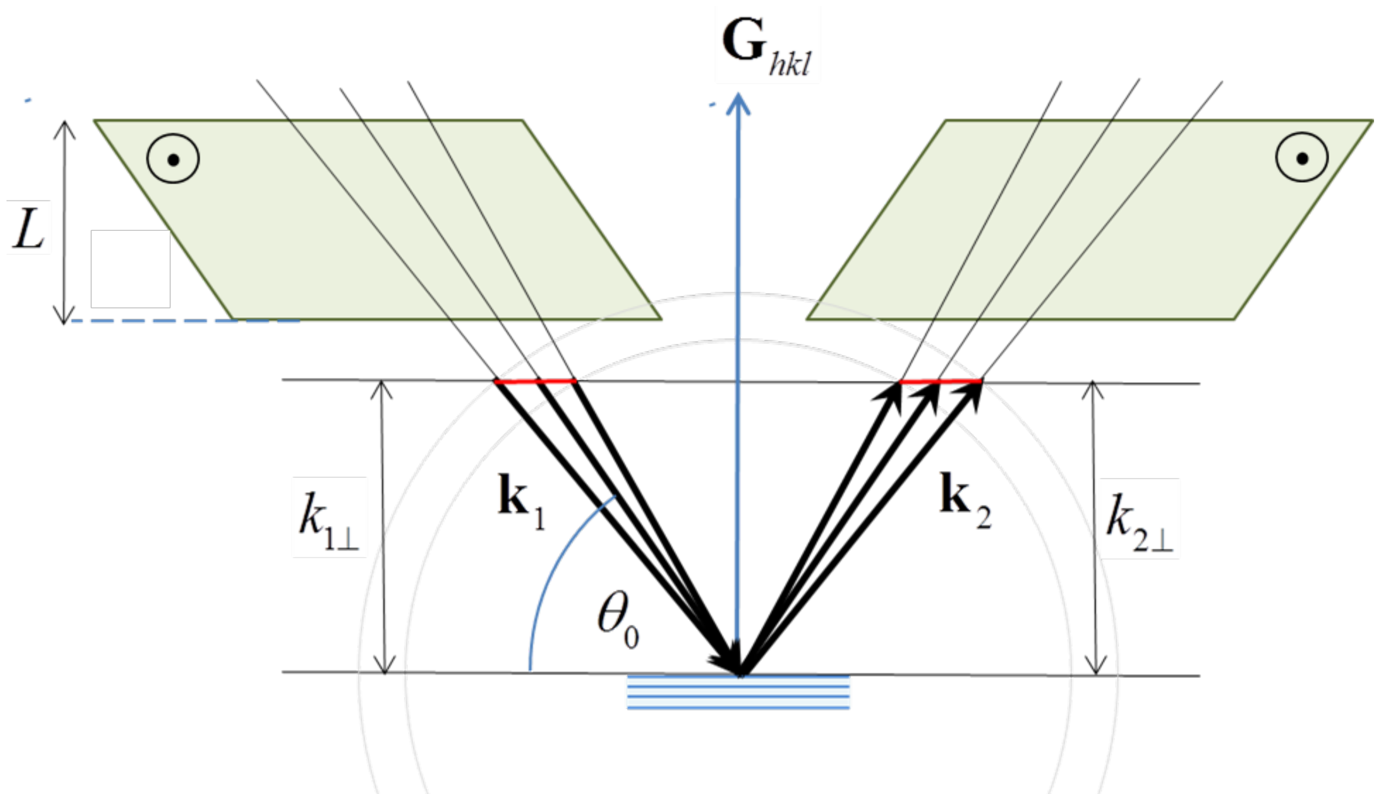


→ accumulated Larmor phase: $\varphi = \omega_L t = \frac{\gamma B l}{v} = \frac{\gamma B L}{v_\perp}$

neutron momentum: $\hbar\mathbf{k} = m\mathbf{v}$, with wave vector $\mathbf{k}$

gyromagnetic ratio $\gamma = \frac{\mu_n}{\hbar/2}$

DOUBLE-ARM LARMOR DIFFRACTION

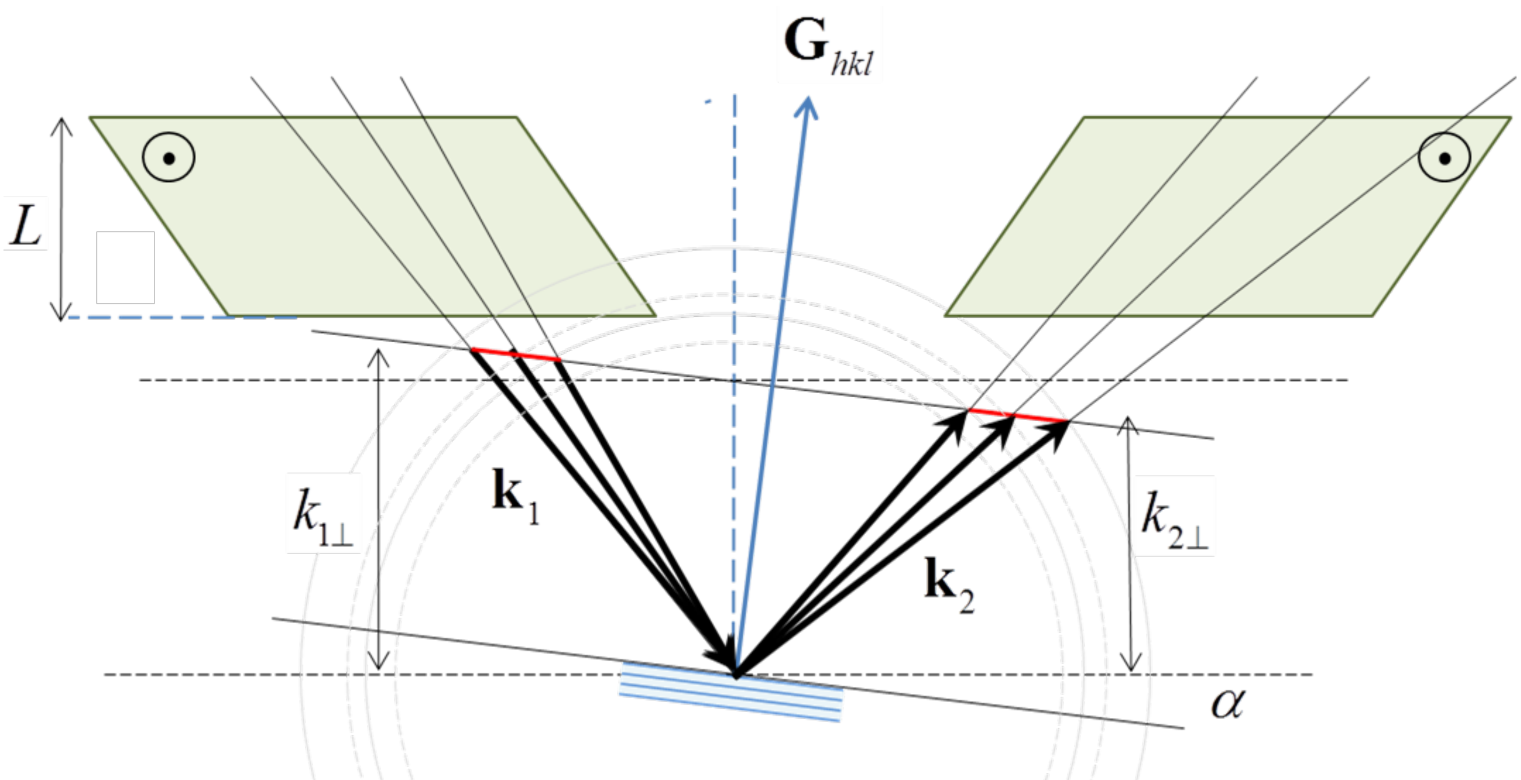


- Consider reflecting planes parallel to face of magnetic field regions
- Larmor phases φ_1 and φ_2 only depend on $v_\perp \propto k_\perp = 0.5 G_{hkl} = \pi / d_{hkl}$
- Independent of beam divergence and monochromaticity !

$$\varphi_t = \varphi_1 + \varphi_2 = 2\pi c B L \left(\frac{1}{k_{1\perp}} + \frac{1}{k_{2\perp}} \right) = 4 c B L d_{hkl}$$

$$c = \frac{\gamma m}{h} = 4.63 \cdot 10^5 \ (\mathrm{Tm})^{-1}\ \mathrm{nm}^{-1}$$

DOUBLE-ARM LARMOR DIFFRACTION

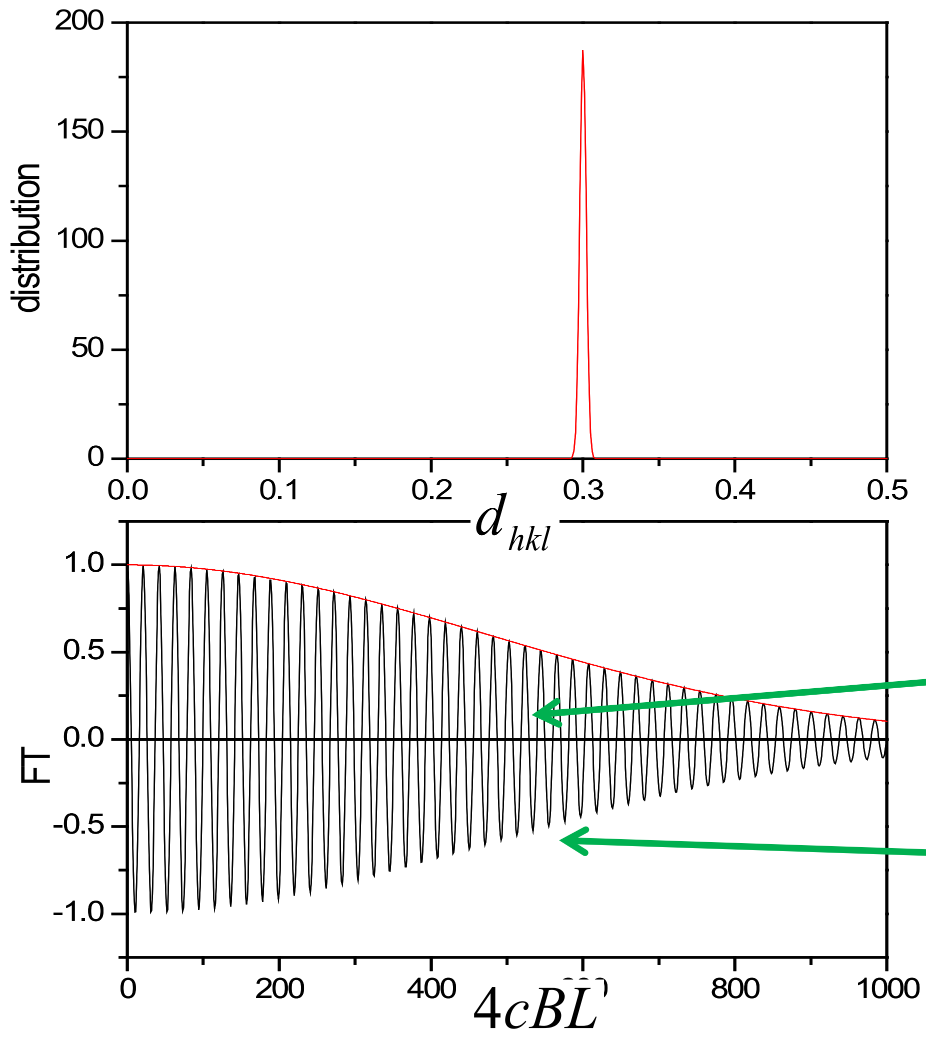


- Misalignment with angle α results in deviation of order α^2

$$\varphi_t = \varphi_1 + \varphi_2 = 2\pi cBL\left(\frac{1}{k_{1\perp}} + \frac{1}{k_{2\perp}}\right) = 4cBLd_{hkl}\left(1 + \frac{1}{2}\left(1 + 2\cot^2\theta_0\right)\alpha^2 + O\left(\alpha^3\right)\right)$$

- For single crystals: resolution determined by misalignment
- For poly-crystals: determined by divergence of incoming and reflected beam

LARMOR DIFFRACTION



With analyser downstream second precession field the intensity measured is

$$I(B) = \frac{1}{2}\left(1 + P(B)\right)$$

$$P(B) = \langle \cos(\varphi) \rangle = \int f(\varphi)\cos(\varphi)\,d\varphi$$

$$\varphi = 4cBLd_{hkl}\left(1 + O\left(\alpha^2\right)\right)$$

period $\rightarrow$
lattice spacing d

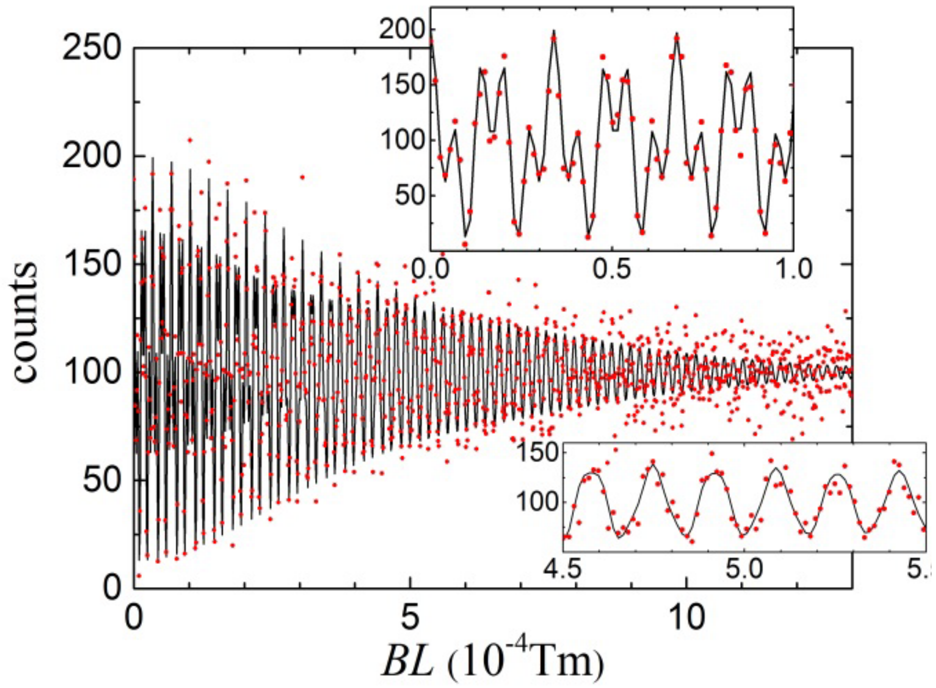
envelope $\rightarrow$
peak shape
multiplied with resolution function

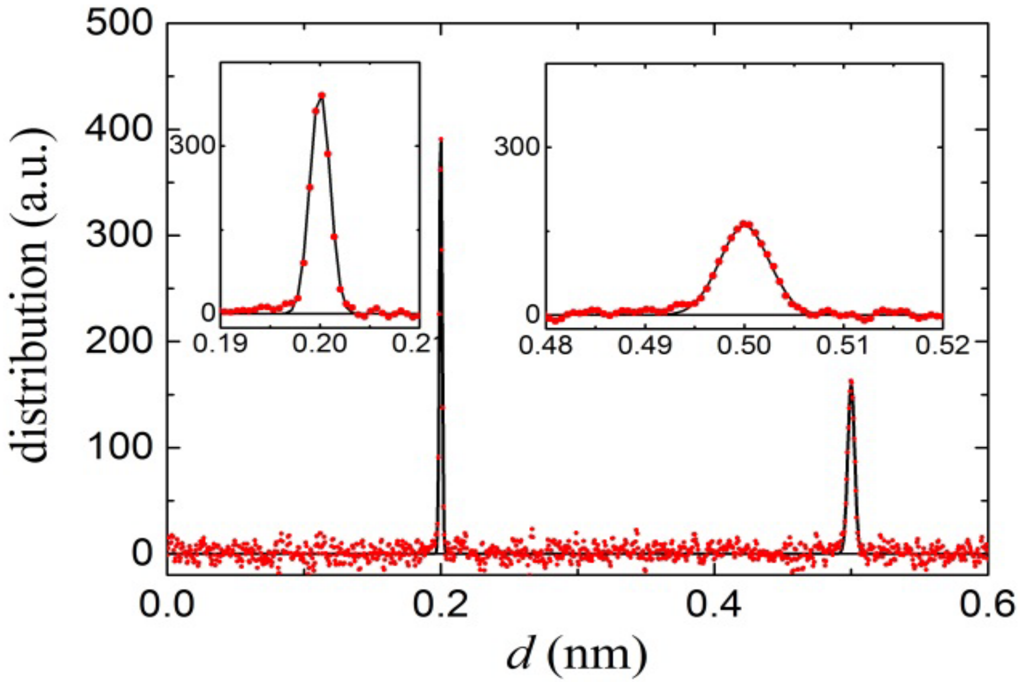
$$\frac{\Delta d}{d} \approx 10^{-6}$$

LARMOR DIFFRACTION

Calculations

- two lattice spacings: d = 0.2 and 0.5 nm
- both with distribution $\Delta d/d$ = 5 10^{-3}





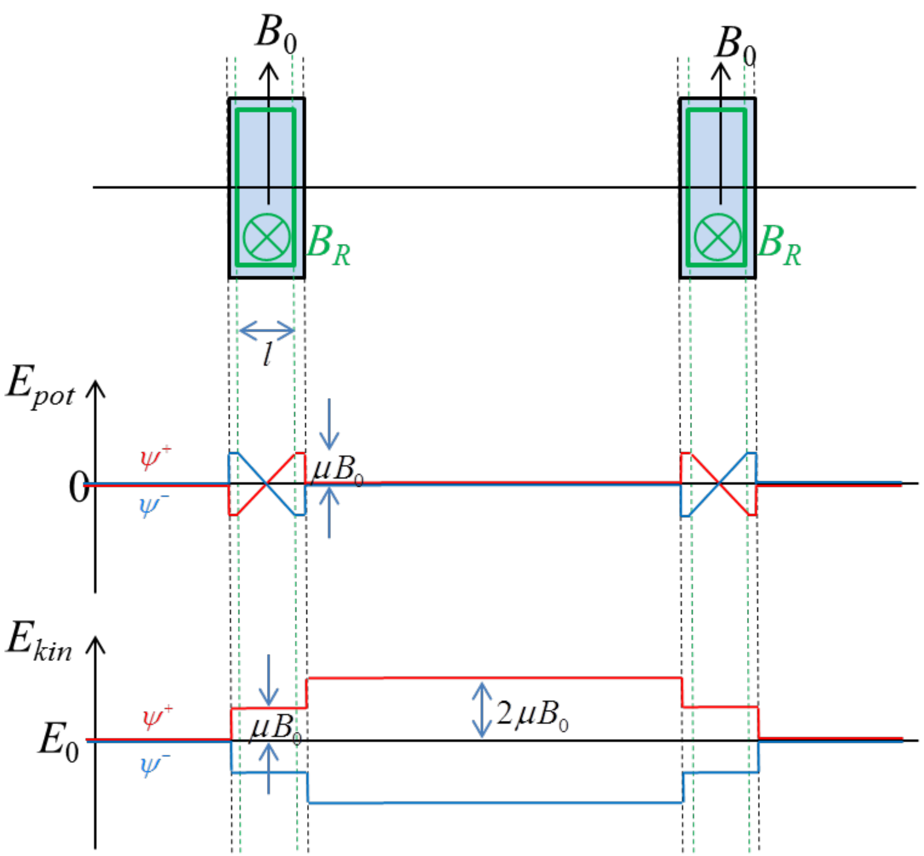
LARMOR DIFFRACTION

Technical realisation

- NRSE (FLEX, HMI; TRISP, FRM2)
- NRSE (prototype, Delft; Offspec/Larmor, ISIS)
- Wollaston prisms (Indiana Univ / ORNL / ISIS)

LARMOR DIFFRACTION
Technical realisation

- NRSE (FLEX, HMI; TRISP, FRM2)

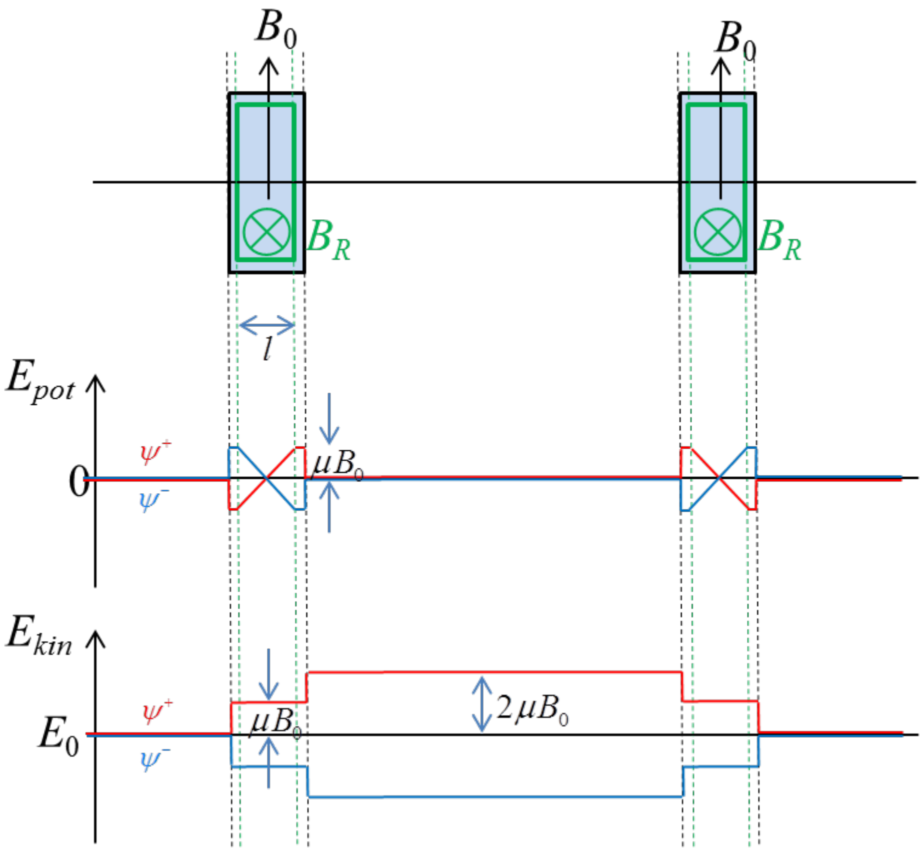


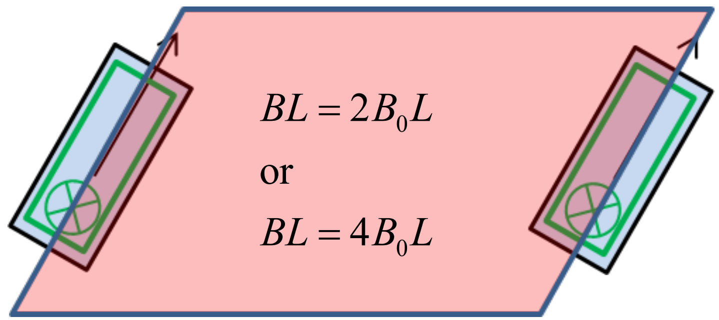
resonance condition: $\omega_R = \gamma B_0$

π-flip condition: $(hm\lambda l)\gamma B_R = \pi$

LARMOR DIFFRACTION
Technical realisation

- NRSE (FLEX, HMI; TRISP, FRM2)





resonance condition: $\omega_R = \gamma B_0$

π-flip condition: $(hm\lambda l)\gamma B_R = \pi$

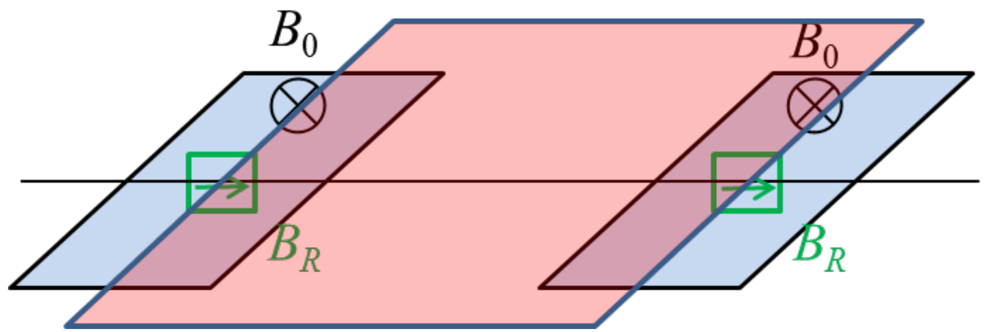
FLEX: first LD experiment

Rekveldt et al., Europhys.Lett. 2001

TRISP: most LD experiments

LARMOR DIFFRACTION
Technical realisation

- NRSE (prototype, Delft; Offspec/Larmor, ISIS)

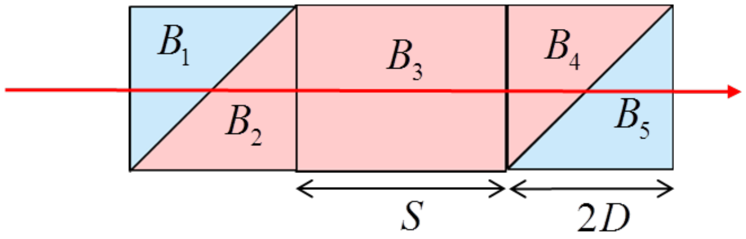


Delft: first single-arm LD experiment
Rekveldt et al., J.Appl.Cryst. 2013
Larmor: first LD experiment: spring 2018

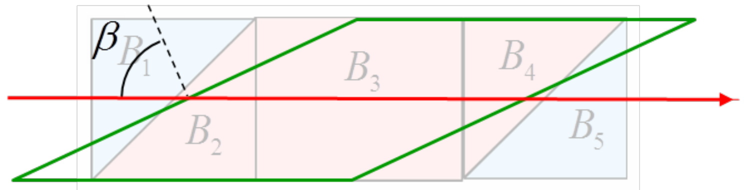
LARMOR DIFFRACTION
Technical realisation

- Wollaston prisms (Indiana Univ / ORNL / ISIS)

Group of Roger Pynn uses a set of triangular superconducting Wollaston prisms to construct a precession region. By choosing the different magnetic fields an effective tilt angle can be set.



$$\tan\beta = -\frac{(S+2D)(B_4-B_5)}{2d(B_4+B_5)+SB_5}$$

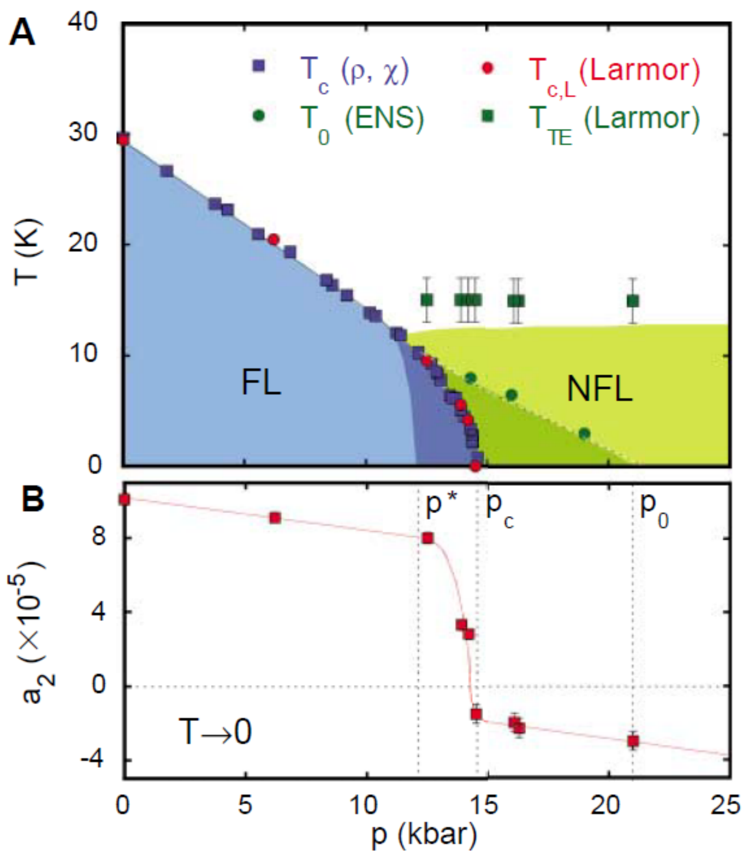


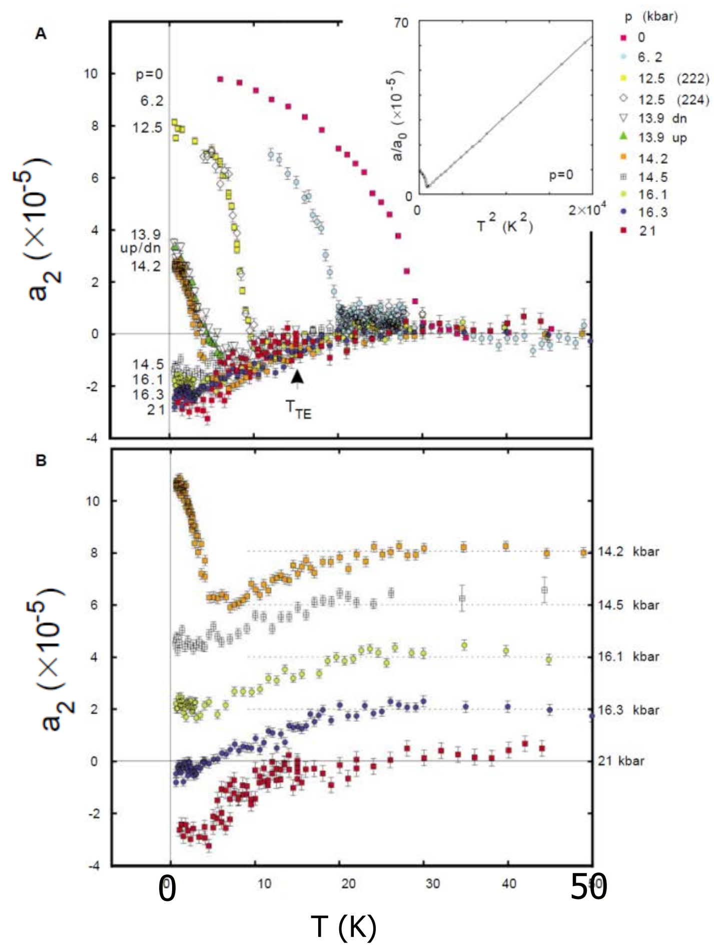
F. Li, R. Pynn, J. Appl. Cryst. **47**, 1849, (2014)

DOUBLE-ARM LARMOR DIFFRACTION

Examples

Fundamental science: single crystal of MnSi





C. Pfleiderer et al, Science **316** (2008) 1510

DOUBLE-ARM LARMOR DIFFRACTION
Examples

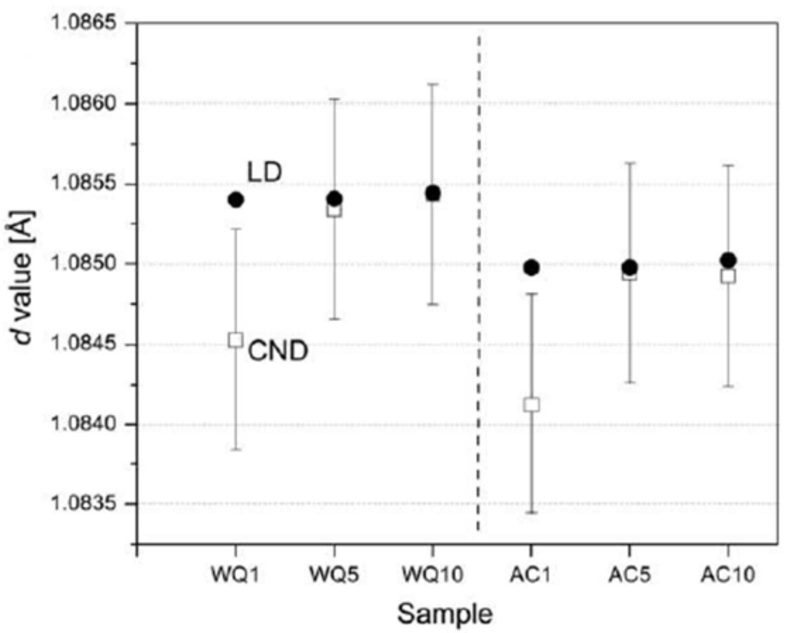
Materials science: different types of Inconel

Absolute lattice spacing

CND: conventional $\Delta d / d = 6.5 \cdot 10^{-4}$

LD: Larmor $\Delta d / d = 1.5 \cdot 10^{-5}$

(accuracy calibration sample)



Fluctuations in d values

single-phase samples:
different size / different heat treatment

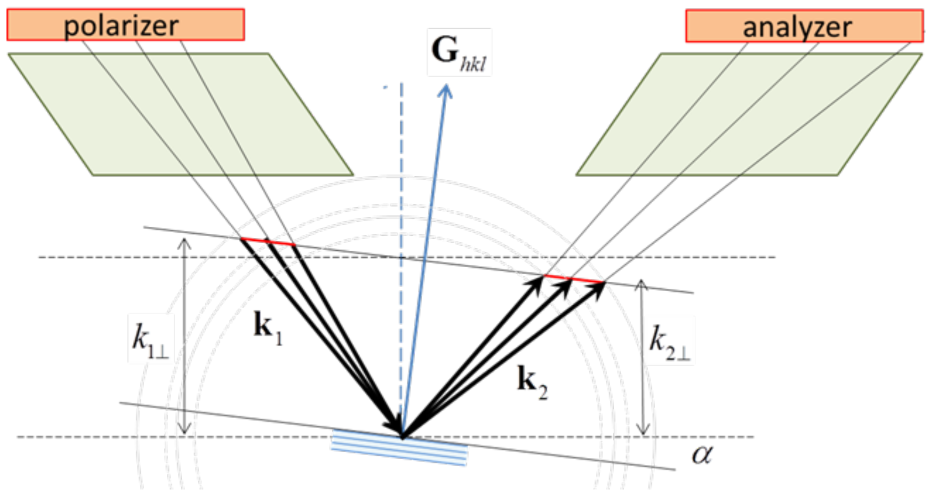
Table 3
Experimental results for the fluctuations of the d values (ε_{FW}) of the γ-matrix phase of single-phase IN 718 samples AC and WQ with different radii (1, 5, 10 mm) observed by LD.

Sample	ε_m
WQ1	1.51×10^{-3}
WQ5	1.55×10^{-3}
WQ10	1.67×10^{-3}
AC1	1.22×10^{-3}
AC5	1.07×10^{-3}
AC10	1.09×10^{-3}

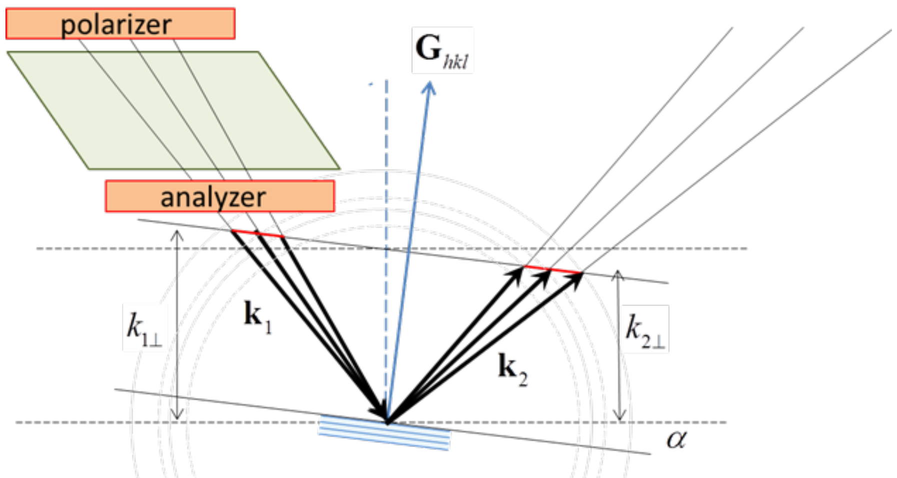
multi-phase samples:
different values for different phases

J. Repper et al, Acta Mater. **58** (2010) 3459

DOUBLE-ARM vs SINGLE-ARM GEOMETRY



Double-arm geometry problematic for magnetic sample / sample environment

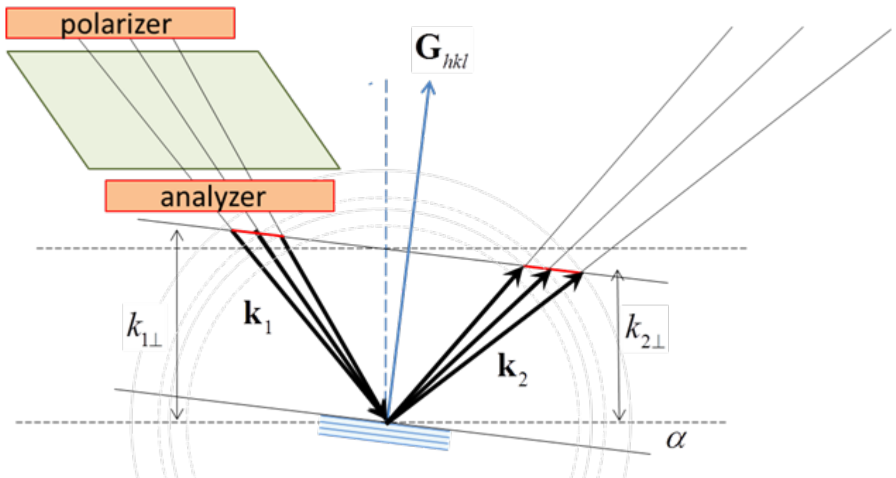


Solution:
Single-arm geometry

Drawback:
No compensation for $k_{1\perp} > k_{2\perp}$

SINGLE-ARM LARMOR DIFFRACTION

Resolution for single crystals



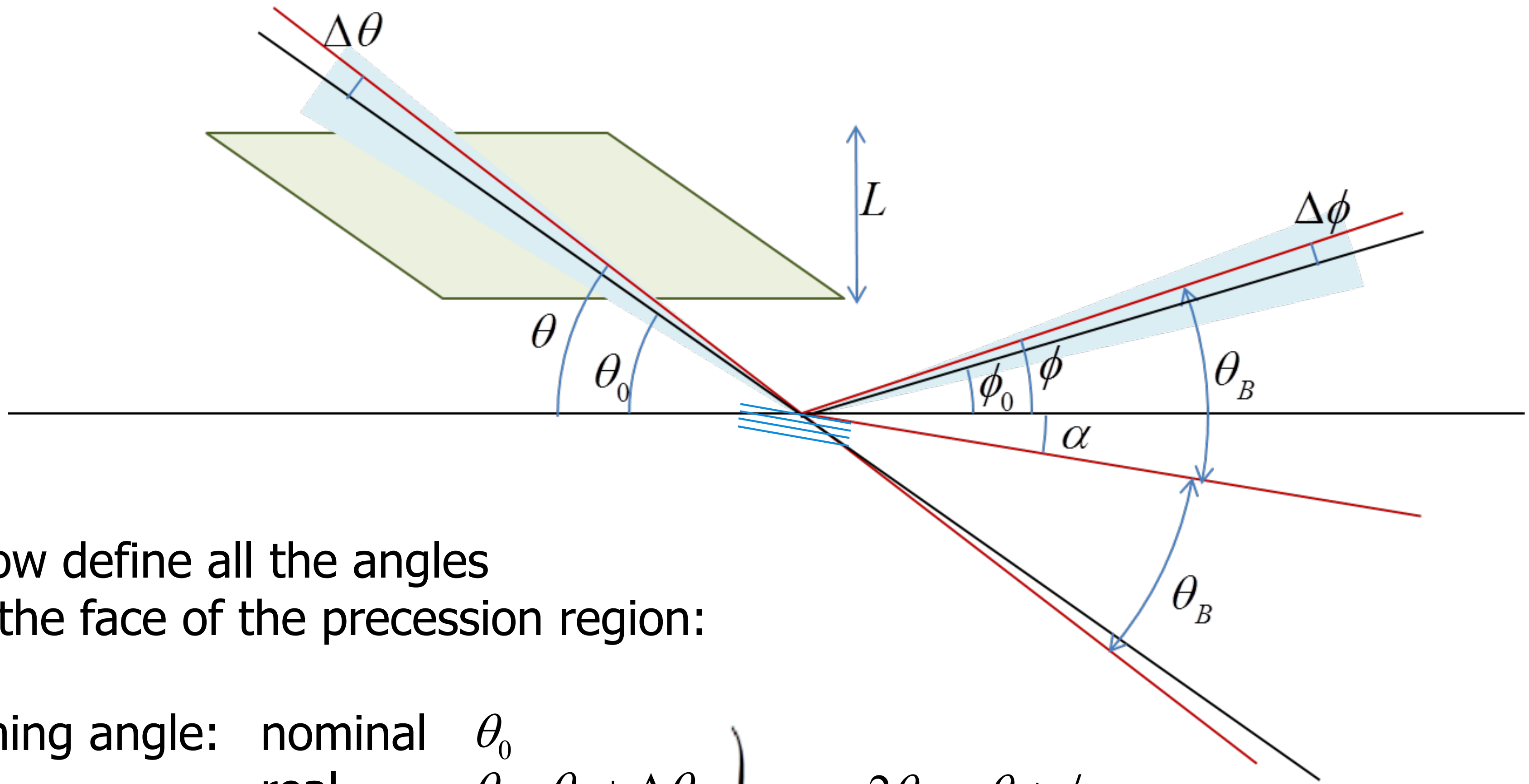
again: align the crystal plane under investigation parallel to face of precession region

$$\varphi_t = \varphi_1 = 2\pi cBL\frac{1}{k_{1\perp}} = 2cBLd_{hkl}\left(1+\alpha\cot\theta_0 + O\left(\alpha^2\right)\right)$$

→ now sensitive to misalignment α in first order

SINGLE-ARM LARMOR DIFFRACTION

Resolution for polycrystalline materials



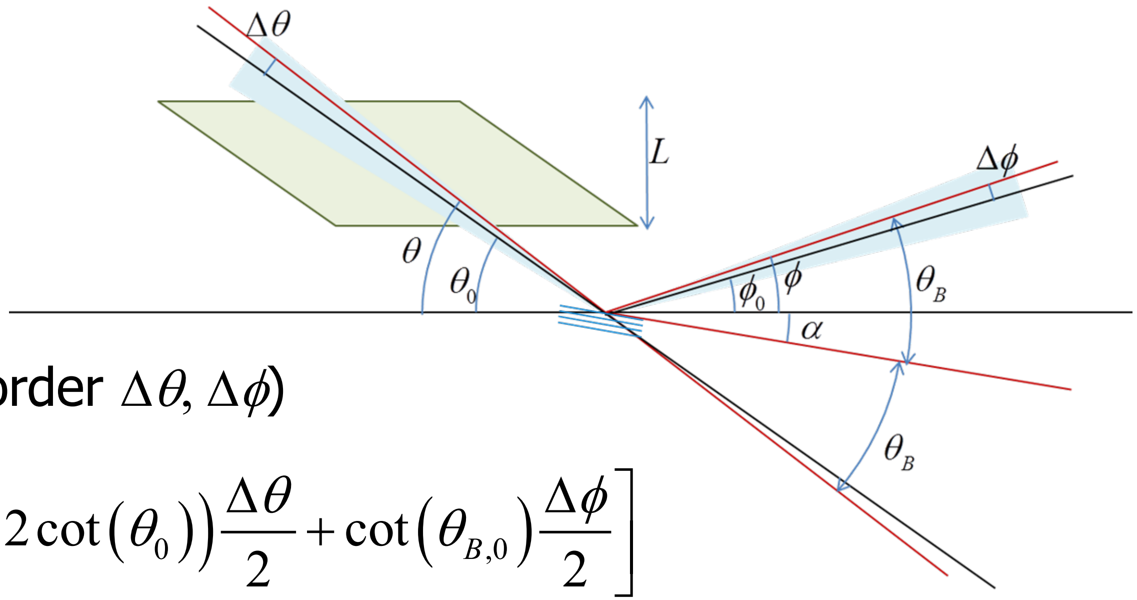
We now define all the angles
w.r.t. the face of the precession region:

Incoming angle:	nominal	θ_0
	real	$\theta = \theta_0 + \Delta\theta$
Reflected angle:	nominal	ϕ_0
	real	$\phi = \phi_0 + \Delta\phi$

$$\rightarrow \quad 2\theta_B = \theta + \phi, \qquad \alpha = \theta_B - \phi$$

SINGLE-ARM LARMOR DIFFRACTION

Resolution poly-crystals



Resulting in a precession angle (1st order $\Delta\theta, \Delta\phi$)

$$\varphi_1 = 2cBLd_{hkl}\frac{\sin(\theta_{B,0})}{\sin(\theta_0)}\left[1+\left(\cot(\theta_{B,0})-2\cot(\theta_0)\right)\frac{\Delta\theta}{2}+\cot(\theta_{B,0})\frac{\Delta\phi}{2}\right]$$

and d-spacing resolution

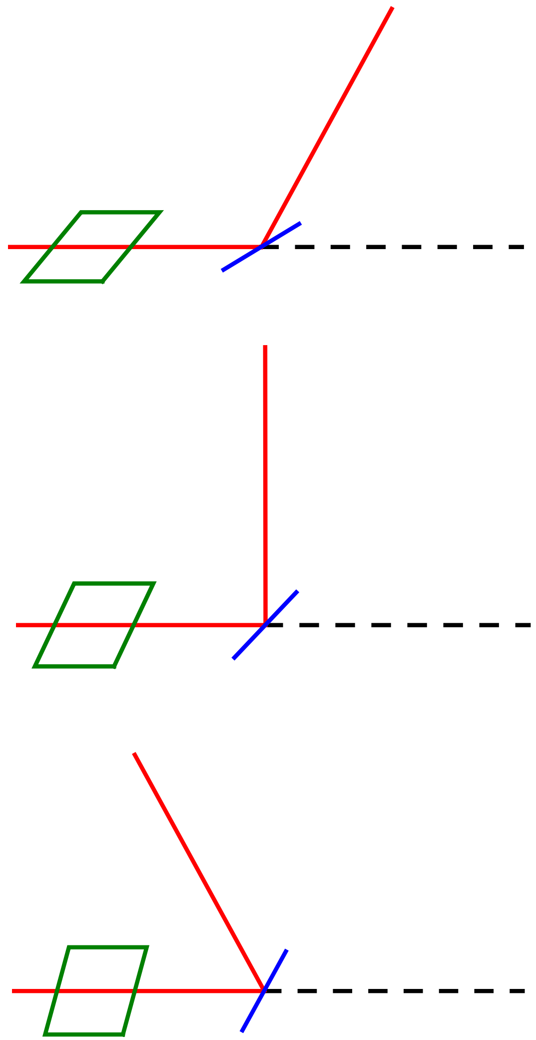
$$\frac{\Delta d}{d} = \frac{1}{2}\sqrt{\left(\cot(\theta_{B,0})-2\cot(\theta_0)\right)^2\Delta\theta^2+\cot^2(\theta_{B,0})\Delta\phi^2}$$

The contribution of the incoming divergence can be cancelled (1st order) by the focussing condition

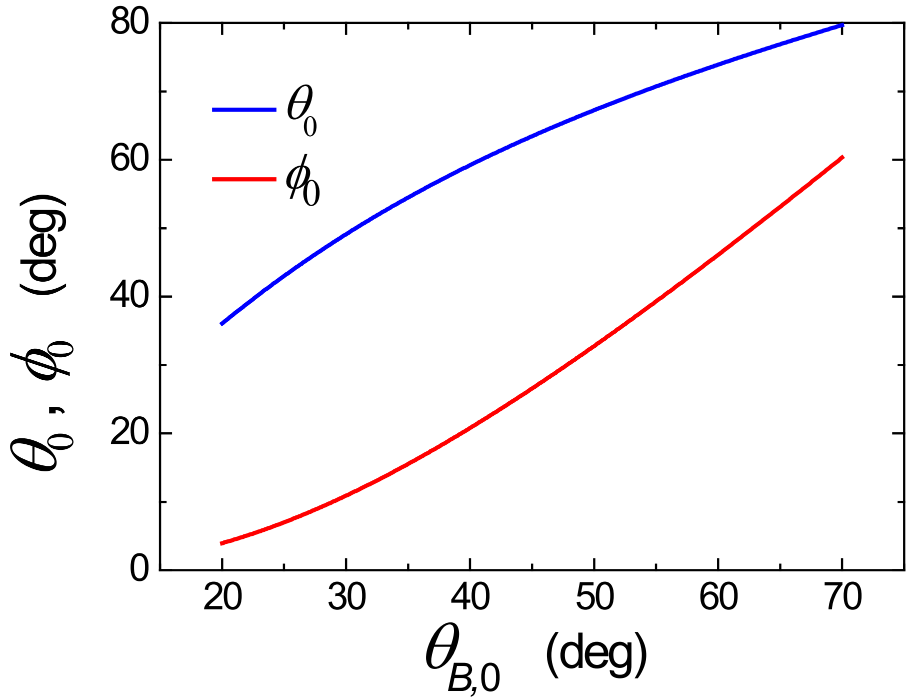
$$\cot(\theta_{B,0}) = 2\cot(\theta_0) \text{ or } \cot\left(\frac{\theta_0+\phi_0}{2}\right) = 2\cot(\theta_0)$$

SINGLE-ARM LARMOR DIFFRACTION

Focussing condition for polycrystalline materials

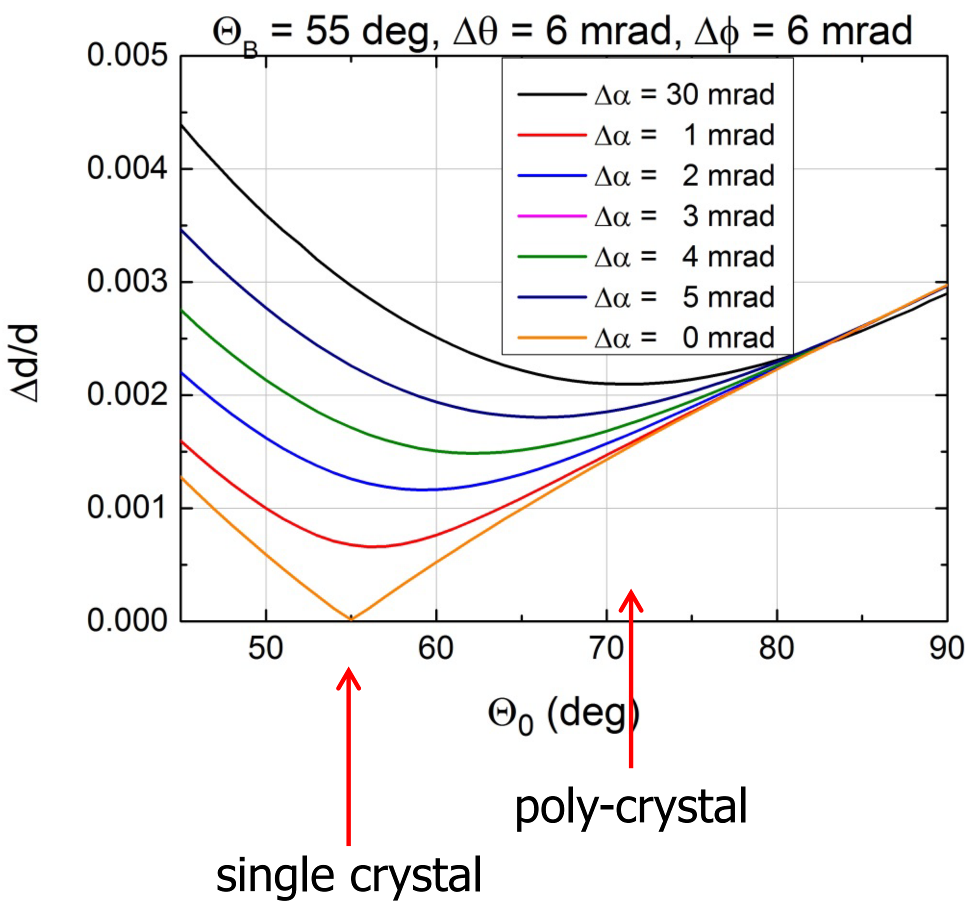


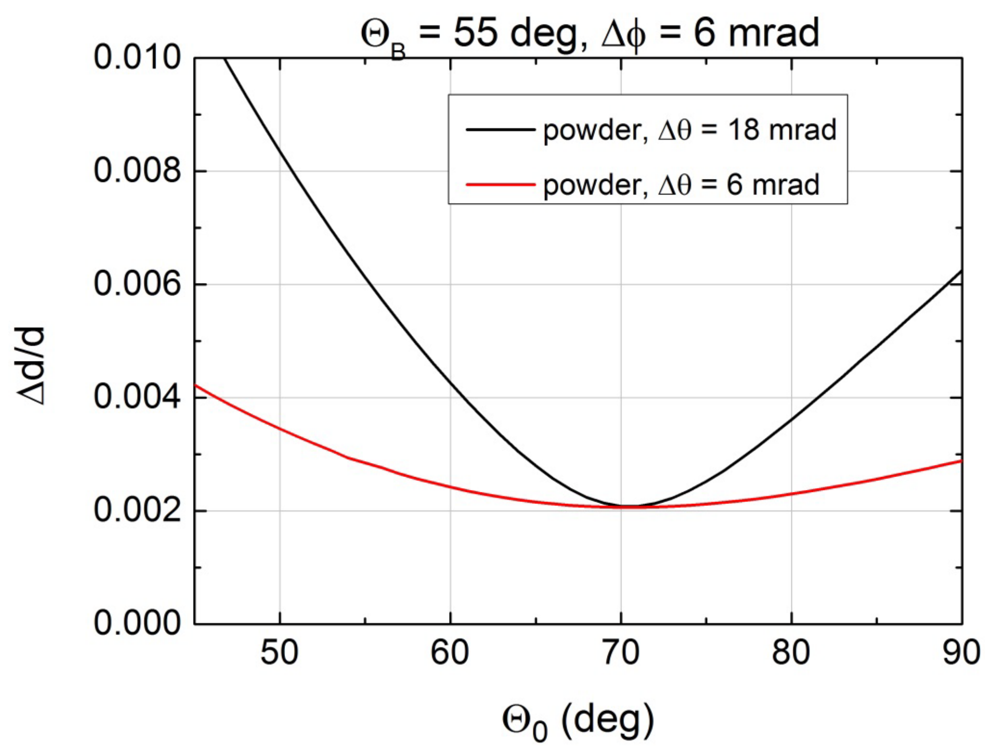
$$\cot(\theta_{B,0}) = 2\cot(\theta_0)$$



SINGLE-ARM LARMOR DIFFRACTION

Focussing condition

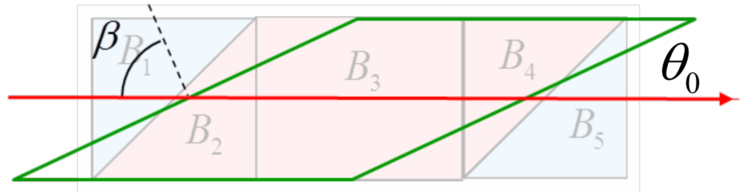




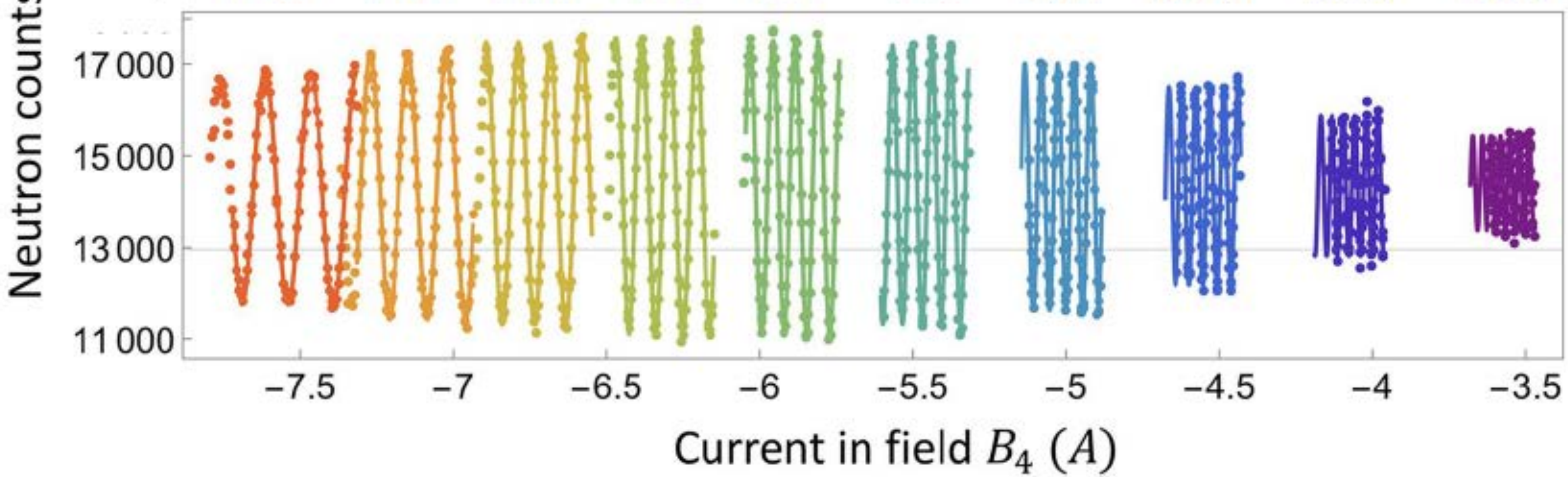
influence divergence of incoming beam

SINGLE-ARM LARMOR DIFFRACTION
Example, Si(111) perfect single crystal

Wollaston prisms, PTAX, HIFR, Oak Ridge
λ = 2.46 Å, θ_B = 23.3°

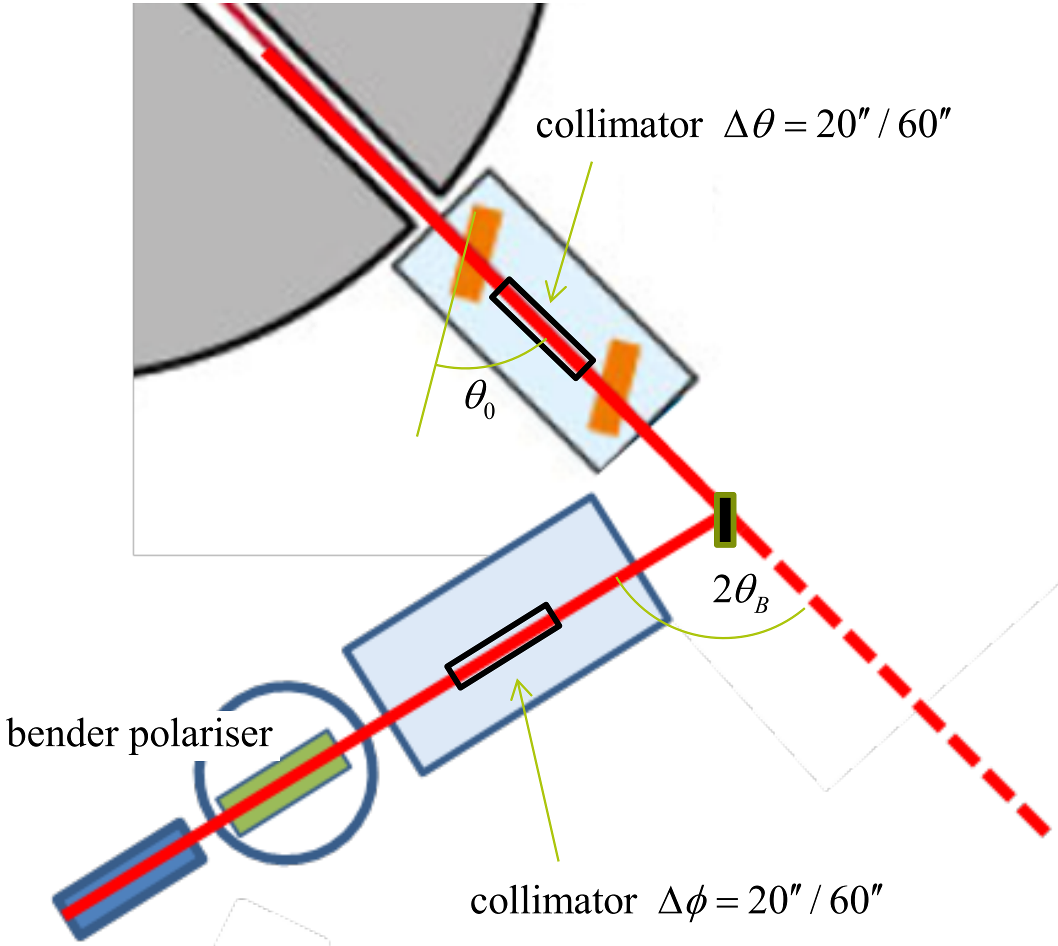


θ_0	15.3	17.8	19.3	21.3	23.3	25.3	27.3	29.3	31.3	33.3
β	74.7	72.7	70.7	68.7	66.7	64.7	62.7	60.7	58.7	56.7



SINGLE-ARM LARMOR DIFFRACTION
Example, Ge(311) perfect / mosaic single crystal

Larmor diffractometer TRISP, FRM2 Munich



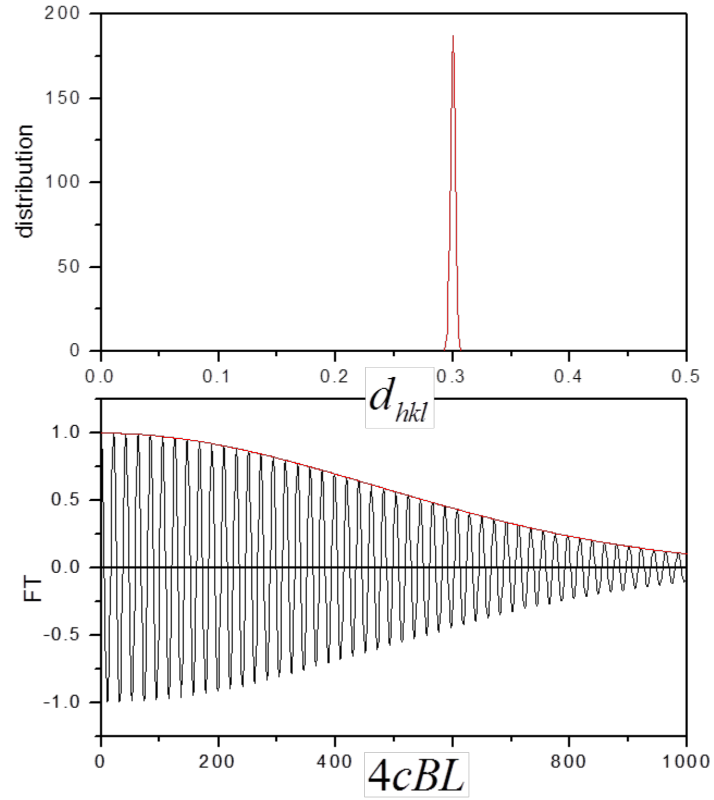
- determine resolution as a function of θ_0

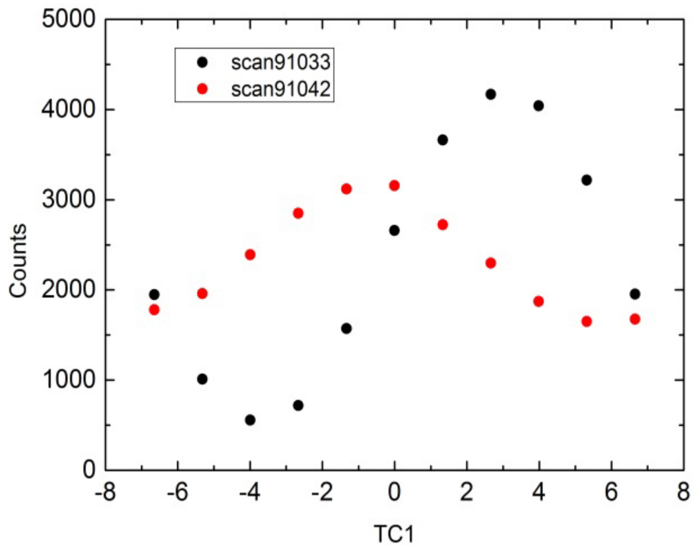
  for fixed Bragg angle θ_B = 55°
  and different divergences
  of incoming and scattered beam

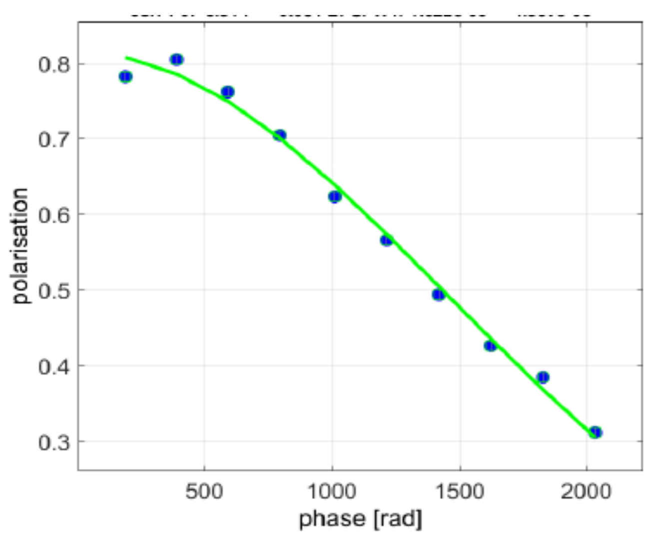
LARMOR DIFFRACTION
Example TRISP

measuring method for examining one Bragg peak:

- 11 BL-point
- BL can be expressed in Larmor phase
- around these points interval covering 1 period → amplitude
- determine damping



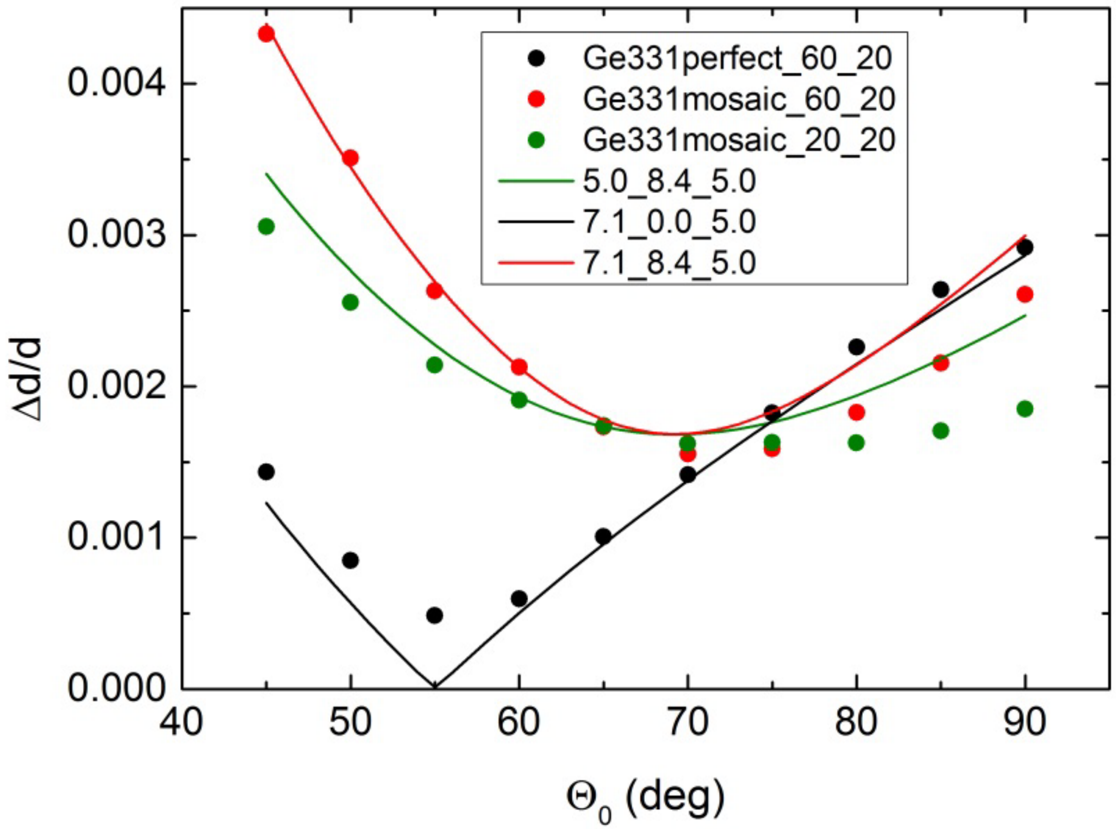




SINGLE-ARM LARMOR DIFFRACTION

Example, Ge(331) perfect / mosaic single crystal

Larmor diffractometer TRISP, FRM2 Munich

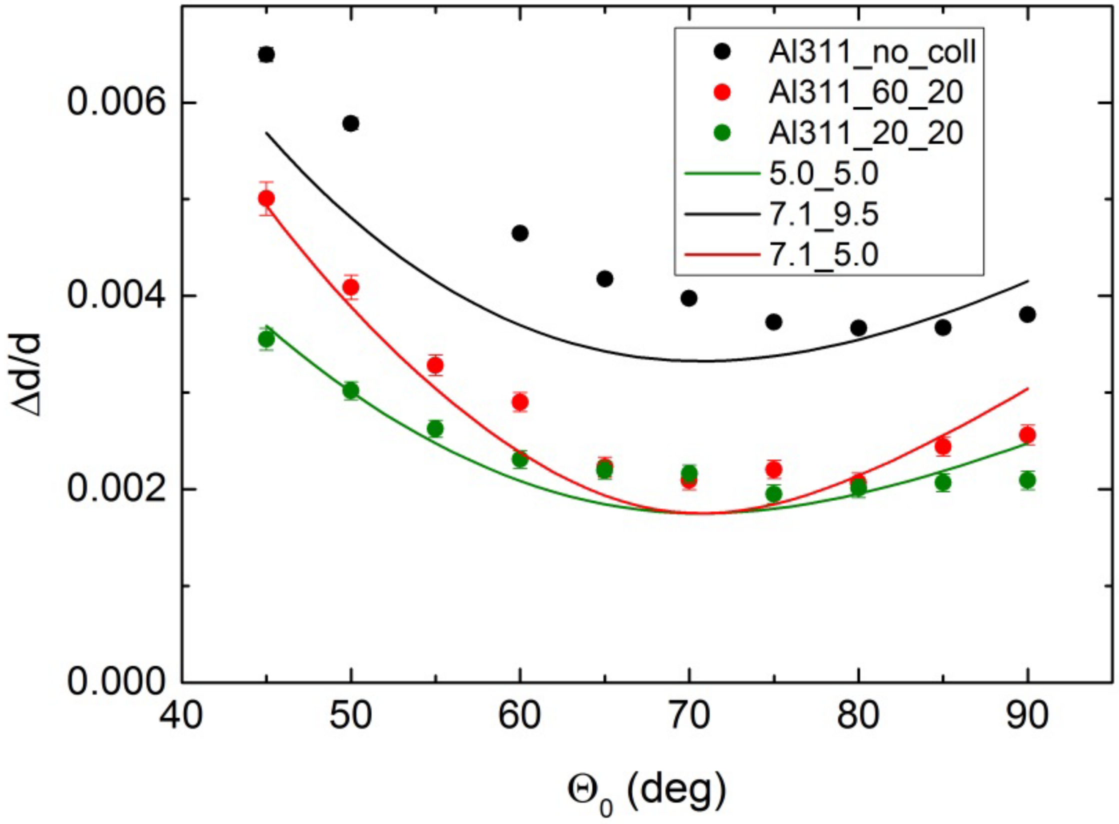


dots: experimental results
black: η=0, $\Delta\theta_{coll}$=18, $\Delta\phi_{coll}$=6 mrad
red: η=8.4, $\Delta\theta_{coll}$=18, $\Delta\phi_{coll}$=6 mrad
green: η=8.4, $\Delta\theta_{coll}$=6, $\Delta\phi_{coll}$=6 mrad

lines: calculations
black: η=0, $\Delta\theta_{coll}$=7.1, $\Delta\phi_{coll}$=5 mrad
red: η=8.4, $\Delta\theta_{coll}$=7.1, $\Delta\phi_{coll}$=5 mrad
green: η=8.4, $\Delta\theta_{coll}$=5, $\Delta\phi_{coll}$=5 mrad

SINGLE-ARM LARMOR DIFFRACTION
Example, Al(311) poly-crystal

Larmor diffractometer TRISP, FRM2 Munich



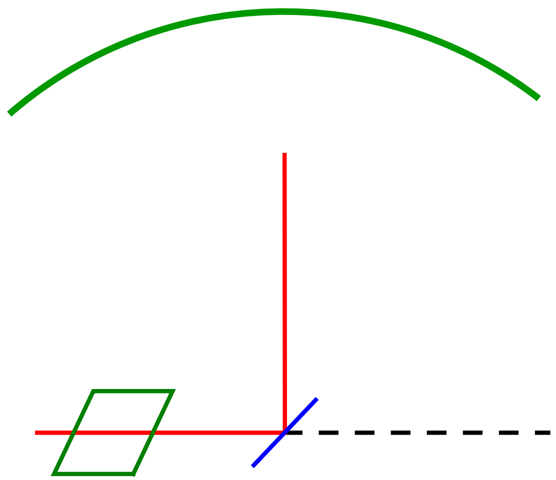
SINGLE-ARM LARMOR DIFFRACTION

- single-crystals: in focussing condition
  resolution sensitive to first order misalignment α
  (for double-arm geometry $\sim \alpha^2$)

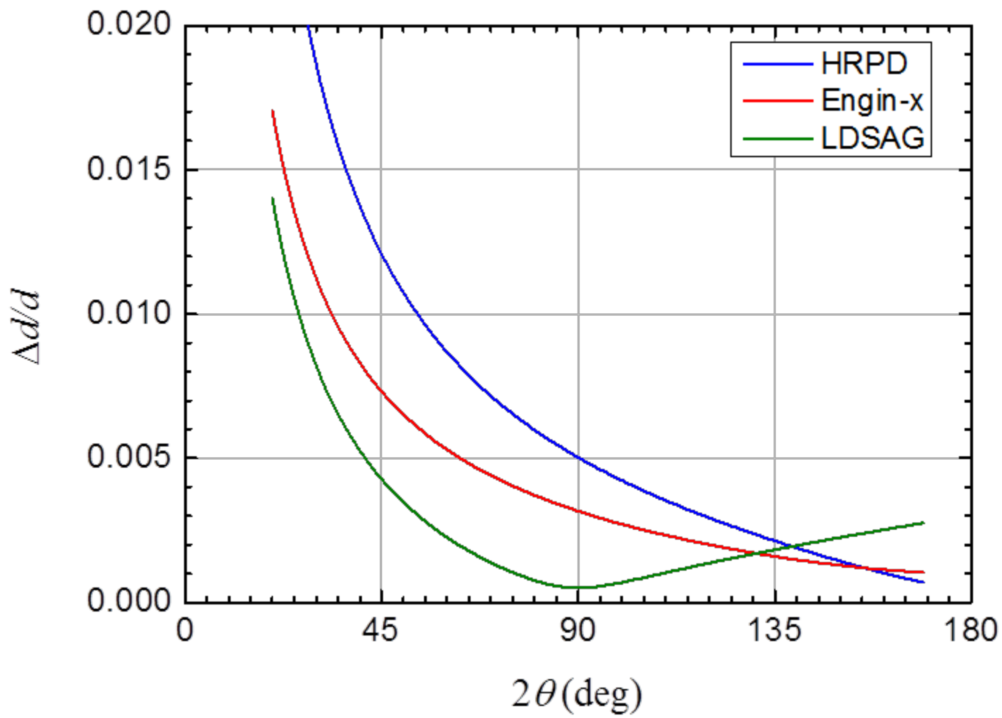
- poly-crystals: in focussing condition
  resolution sensitive to first order divergence scattered beam $\Delta\phi$
  (can be made small by small sample diameter and detector-pixel size)
  resolution sensitive to second order divergence incoming beam $\Delta\theta^2$

- if precession region only in scattered beam
  focussing condition cancels first order divergence scattered beam

SINGLE-ARM LARMOR DIFFRACTION
comparing resolution



$\theta_0 = 63.4^{\circ}$



- Engin-x values for divergences are used, i.e. $\Delta\theta = 0.006$, $\Delta\phi = 0.002$ (distance sample-detector 1.5 m, 3 mm sample, 3 mm detector pixel)
- Note that Larmor diffraction is not sensitive for time-of-flight resolution $\Delta t/t$

DISCUSSION

- The Larmor diffraction (LD) technique, especially double-arm LD, has extremely good resolution, down to $\Delta d/d \approx 10^{-6}$

- For magnetic samples single-arm LD is a good alternative. The resolution is not as good as LD-DAG, but better than conventional neutron diffractometers.

- LD is a Fourier technique: well-suited for studying one or a few Bragg peaks, not suited to dissolve many Bragg peaks, in particular the low-intensity peaks. Similar to Fourier Chopper – RTOF technique (e.g. FSS GKSS), but with better resolution and no high-speed mechanical components.

- Comparison LD at reactor source $\leftrightarrow$ spallation source:
  - Since the sample acts as a monochromator the intensity for the wavelength probed, will in general be much higher for a reactor source (not for ESS!)
  - on the other hand: at a spallation source many Bragg peaks are probed simultaneously, in different tof-intervals.

    $\rightarrow$ = in 2-phase system: precise d-spacing and distribution for both phases
    = Q-dependent line broadening: grain size $\leftrightarrow$ micro-strain contributions
    = more lattice planes $\rightarrow$ info about anisotropic strain
    = separate two overlapping Bragg peaks

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